

Responses to Referee #2 Comments

General comment: The authors' have presented temporal variations in the mass concentrations of carbonaceous and inorganic constituents in the tropical Indian aerosol (PM₁₀) collected on day- and night-time bases during winter (23 January to 6 February) and summer (22–31 May) of year 2007 from Chennai, India. Overall, the paper is well written and provides an interesting data set on carbonaceous and inorganic aerosols. However, the text and discussion of results require further clarification. Perhaps, it is not appropriate to state in the abstract that samples collected from tropical site in India (Chennai, 13.04 °N, 80.17 °E) can better characterise south and southeast-Asian aerosols. In the same context, it is incorrect to state in the MS title as “tropical Indian aerosols”. It may rather be referred as tropical aerosols from Indian region. There are some studies reported from tropical climate in western India. The paper may be published in ACP after a minor revision.

Response: The authors appreciate the reviewer's comments on this manuscript. Following the reviewer's suggestion, we modified these phrases in the revised manuscript (please see page 1, line 2 and page 2, line 14 & 15).

Major comment: The variability in the mass concentrations of carbonaceous and inorganic aerosols has been attributed to the origin of air masses and their source strength. Although day- and night-time variability in the mass concentrations of chemical constituents is an important outcome of this work; authors may like to compare their data set from other sites in the context of Indian aerosols. Also, authors should discuss WSOC/OC and WIOC/EC ratios with other studies from Indian region.

Response: Following the reviewer's suggestion, we discussed only the WSOC/OC ratios in Chennai in comparison with those from other studies in Indian region in the revised MS (please see page 19, lines 448–456). But we did not discuss the WIOC/EC ratios as they are not available in other studies from Indian region to the best of the author's knowledge.

Specific comments: P3941, Section 2.1: Please discuss day-night variability in aerosol constituents.

Response: We did not think that it is important to discuss day and night variability in aerosol constituents in section 2.1. However, we discussed the possible day- and night-time variability in aerosol constituents at Chennai due to land/sea breeze circulation in the revised MS (please see page 5, lines 103–113), following the reviewer's suggestion.

P3942, L5: The analytical errors for duplicate analysis: 1.2% (for OC) and 1.7% (for EC) are very low. What are the detection limits for these two constituents?

Response: Yes, the analytical errors obtained in duplicate analysis of our samples are lower than the specified range (4–6%) by Sunset Laboratories. The detection limits for both EC and OC are 0.2 µg cm⁻². The detection limits are now added in the revised MS (please see page 7, lines 134 & 135).

P3942, L11: "TC" needs to be spelled out in the early text.

Response: We gave the full name of TC in the revised MS (please see page 7, line 142).

P3943, L4–5: How the authors know that aerosols in Chennai (mainland) are affected by wood smoke a priori?

Response: As evident from the literature, anthropogenic (biofuel/biomass burning) emissions are significant in India as well as in the region. However, we modified the phrase in the revised MS (please see page 8, line 163).

P3943, L12–13: What are the minimum OC/EC ratios used for the estimation of SOC in summer and winter?

Response: The minimum OC/EC ratio was 0.7 in winter and 3.35 in summer. We included these ratios in the revised MS (please see page 8, line 178).

Section 3.1: Simultaneous discussion of carbonaceous and inorganic aerosols is often confusing in the text. This section requires reorganisation.

Response: Following the reviewer's suggestion, we re-organized the section 3.1 by dividing it into three sub-sections in the revised MS (please see page 10–13, lines 210–304).

P3946, L 5-7: Average equivalent ratios of total cations to anions are 0.85 and 1.21 in winter and summer respectively; suggest that aerosols are more acidic in winter than summer. Authors attribute the acidity due to H^+ ions (not measured in this study) in aerosols. Could this be attributed to the seasonal variability in organic acids in aerosol samples? Authors may like to address these points in greater detail.

Response: As we did not measure the pH, it is not appropriate to attribute the acidity to organic acids. However, we noted the concentrations of diacids and possible association of cations with organic acids in Chennai aerosols in the revised MS (please see page 13, lines 297 & 298 and 301 & 302).

P3946, L 10-12: NH_4^+/SO_4^{2-} equivalent ratios are much lower in Chennai aerosols (0.63 in winter and 0.81 in summer) compared to those over other Indian regions (as per the cited references). What could be the reasons? The possible association of acidic species with mineral dust (Ca^{2+} and Mg^{2+}) need to be addressed.

Response: No, NH_4^+/SO_4^{2-} equivalent ratios found in Chennai aerosols are close to those from some sites like Agra (ca. 0.83; Parmar et al., 2001) in India and those influenced by biomass burning emissions like Himalayas (ca. 0.8; Carrico et al., 2003).

We believe that it is not important to extend the discussion on neutralization of acidic species in association with mineral species (Ca^{2+} and Mg^{2+}) because the NH_4^+/SO_4^{2-} molar ratios in Chennai aerosols were always more than 1 (equivalent ratios range: 0.67-1.58; $n = 49$) that indicate the sufficiency of NH_3 to neutralize most of the acidic species forming NH_4HSO_4 preferably rather than $(NH_4)_2SO_4$, and NH_4NO_3 and NH_4Cl . On the other hand, the concentrations of Ca^{2+} and Mg^{2+} were very low in Chennai aerosols (please see Table 1). This point is included in the revised MS (please see page 13, lines 302-304).

P3946, L 15-16 and P3591, L9-11: Higher concentrations of EC and some ionic species have been attributed to the origin of air masses and their source strength. What are the representative OC/EC ratios for different air masses?

Response: It is difficult to provide the representative OC/EC ratios for different air masses because OC/EC ratios show a broad range (0.6-4.2) depending on type of biofuel and burn rate (Stone et al., 2010). This range is included in the revised MS (please see page 11, lines 236-238).

P3946, L 17-18: Additional sources of organic aerosols in summer are discussed except secondary formation. These could be important sources of OC in summer as secondary aerosol formation could contribute to OC but not to EC. This may also be reflected in OC/EC, WIOC/EC and WSOC/OC ratios in winter and summer.

Response: WSOC/OC ratios in Chennai aerosols are highly comparable between winter and summer, which suggest that the secondary production of organic aerosols may be equally significant in both winter and summer. In fact, concentrations of total diacids (signature compounds for secondary production) were higher in winter than summer in Chennai aerosols (Pavuluri et al., 2010), suggesting that source and/or source strength is more important in varying the concentrations of Chennai organic aerosols seasonally rather than secondary production. Hence, we decided not to add more discussion about this because it is not necessary important to discuss the secondary production in the discussion section on seasonal variations of carbonaceous components in this study.

Section 3.3: The day- and night-time variability in both winter and summer has been discussed for various chemical constituents. Authors may like to address on the formation of secondary inorganic aerosols. It is surprising to note that K^+ concentration (biomass burning tracer) higher during daytime. This observation needs better explanation.

Response: As discussed in section 3.3 in the discussion paper (section 3.5 in the revised MS), sea breeze causes to increase the concentration of K^+ in daytime by uplifting the aerosols transported offshore in previous night and accumulated over oceanic region that enriched with primary pollutants like K^+ . In addition, local emissions from biofuel burning in daytime might have also been influenced the concentration of K^+ for some extent. This point is included in the revised MS (please see page 25, lines 588 & 589).

P3946, L 11-12: What evidence authors may like to provide for SO_4^{2-} and MSA^- production from biomass burning emission?

Response: We have improved the discussion on the source of SO_4^{2-} , in particular, based on its seasonal distribution in the revised MS (please see page 12, lines 268-278). From the literature, it is evident that biomass burning emits dimethyl disulfide and dimethyl sulfide; the precursors of SO_2 and MSA (Meinardi et al., 2003). Seasonal distributions of ionic species together with air mass trajectories imply that biomass burning is the major source of SO_4^{2-} and MSA^- in Chennai aerosols although we could not preclude the contributions from fossil fuel combustion and marine sources for some extent. Please see page 12, lines 268-287 in the revised MS.

P3951, L18-19: The OC/EC ratios are characteristically different in summer (6.2) and winter (1.6) (Table 2). A large temporal change can not be attributed to SOA formation alone. A close similarity in WSOC/OC ratios in summer and winter; P3953, L26) further supports this fact. Authors needs to consider that WIOC/EC ratios in winter and summer (0.8 and 3.6, respectively; P3954, L21-22) are quite different; suggesting that sources could be different in summer and winter.

Response: Yes, we agree with the reviewer that the large temporal changes in concentration were mainly due to differences in types of sources (biofuel/biomass) in winter and summer. This point is included in the revised MS (please see page 17, lines 394-397).

P3953, L26: WSOC/OC ratios for day- and night-time samples can be stated as it is not clear from the Fig. 6. Why authors compare WSOC/OC and WIOC/EC ratios over the world but not from Indian regions. A separate discussion on the WSOC/OC ratios over Indian regions should be added in the text.

Response: Following the reviewer's suggestion, we included WSOC/OC ratios for day- and night-time in the text in the revised MS (please see page 18, lines 426 & 427). We discussed the WSOC/OC ratios in comparison with other sites over the world in the context of Indian aerosols. However, we included the discussion on WSOC/OC ratios in Chennai in comparison with other sites from Indian region in the revised MS (please see pages 19, lines 448-456).

Table 2: Data for Manora Peak and Mt Abu needs to be checked for EC and OC concentrations.

Response: There were typos in Table 2 for Manora Peak and Mt. Abu data. We corrected them in the revised MS.